

VISCOSITY AND SURFACE TENSION OF SATURATED TOLUENE
FROM SURFACE LIGHT SCATTERING (SLS)¹

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ABSTRACT

It is demonstrated that dynamic light scattering (DLS) on a horizontal gas-liquid interface can be used for the reliable determination of surface tension and liquid kinematic viscosity. In contrast to the more usual approaches of surface light scattering (SLS) spectroscopy a set-up is used and described here which makes it possible to measure the capillary wave propagation characteristics in forward scattering direction at variable wave numbers. The experiments in this work rely on a heterodyne detection scheme and signal analysis by photon correlation spectroscopy (PCS). Surface tension and liquid viscosity data of the important and thus well documented reference fluid toluene have been measured under saturation conditions over a wide temperature range from 263 K to 383 K. These data demonstrate the excellent performance of the surface light scattering technique. The achievable accuracy of this technique is discussed in detail for both properties in connection with reference values available in the literature.

KEY WORDS: dynamic light scattering; surface light scattering; surface tension; toluene; viscosity.

1. INTRODUCTION

Light scattering by thermally excited capillary waves on liquid surfaces or gas-liquid interfaces can be used for the investigation of viscoelastic properties of fluids. Based on early theoretical and experimental work [1-10] renewed interest in surface light scattering (SLS) in recent years has resulted in a number of new applications [11-20]. Objects of these investigation extend from simple fluids over polymer solutions, liquid crystals, surfactant monolayers, supramolecular systems to high-temperature melts. The determination of the thermophysical properties surface tension and viscosity is of special interest for pure fluids and fluid mixtures. In this context the SLS method is well established for the investigation of the critical behavior of the surface tension [5, 6, 8, 10], while in general for the viscosity due to instrumental broadening effects only a poor accuracy achievable is reported in literature. At the present time, the application of the surface light scattering technique to simple fluid surfaces serves rather as a check of the somewhat complex calibration procedures involved with this technique [21].

In contrast, in the present work it is demonstrated that the SLS method can be used for a reliable determination of surface tension and viscosity - without any calibration procedure - with an accuracy comparable or even better than for conventional methods. After an introduction into the technique the experimental set-up is described, which allows the analysis of scattered light in forward direction at relatively high and variable wave numbers of capillary waves. In the second part of the paper, the data evaluation procedure is introduced, and results for the surface tension and the liquid kinematic viscosity of toluene are discussed in

comparison with literature. Data for this important reference fluid are obtained under saturation conditions and cover a temperature range from 263 to 383 K.

2. PRINCIPLE OF SURFACE LIGHT SCATTERING (SLS)

2.1 Surface Fluctuations

Liquid surfaces in macroscopic thermal equilibrium exhibit surface waves that are caused by the thermal motion of molecules and that are quantised in so-called “ripples” [1]. Based on a classical hydrodynamic approach thermally excited surface fluctuations result in typical amplitudes of about 10 nm and wavelengths of about 10 μm [2, 3]. In order to excite surface fluctuations work has to be done against the forces acting on a liquid surface. Due to the typically small values of the wavelengths and amplitudes capillary forces dominate, while gravitational forces can be neglected [10].

In general, for the temporal decay of surface fluctuations two cases may be distinguished. In the case of large viscosity and / or small surface tension the amplitude of surface waves is damped exponentially, while in the case of small viscosity the amplitude decays in form of a damped oscillation as it is relevant in this work. Therefore, in the following, an overdamped behaviour of surface fluctuations is not discussed.

A thermally excited surface can be represented by a superposition of waves with different amplitudes \mathbf{x}_q and wave vectors \vec{q} [10]. For a particular surface mode with frequency ω the time-dependent vertical displacement \mathbf{x} of the surface to its flat equilibrium state at a given point \vec{r} is given by

$$\mathbf{x}(\vec{r}, t) = \mathbf{x}_q \exp[i(\vec{q}\vec{r} + \mathbf{a}t)]. \quad (1)$$

For the propagation of capillary waves on a vapor-liquid interface the complex frequency \mathbf{a} of a certain surface mode can be represented in first order approximation by

$$\mathbf{a} = \mathbf{w} + i\mathbf{G} \approx \left[\frac{\mathbf{S}q^3}{\mathbf{r}' + \mathbf{r}''} \right]^{1/2} + i 2q^2 \frac{\mathbf{h}' + \mathbf{h}''}{\mathbf{r}' + \mathbf{r}''}, \quad (2)$$

where \mathbf{S} is the surface tension, \mathbf{r}' and \mathbf{r}'' are the densities of the liquid phase and the vapor phase, respectively, \mathbf{h}' and \mathbf{h}'' are the dynamic viscosities of the liquid and the vapor phase, respectively. Furthermore, the real part in Eq. (2) represents the frequency \mathbf{w} and the imaginary part the damping \mathbf{G} of the surface mode. An exact description of the dynamics of surface waves dependent on surface tension, viscosity and density of the liquid and vapor phase, and the wave vector is obtainable by solving the dispersion equation

$$\left[i\mathbf{a} + 2q^2 \frac{\mathbf{h}' + \mathbf{h}''}{\mathbf{r}' + \mathbf{r}''} \right]^2 + \frac{\mathbf{S}q^3}{\mathbf{r}' + \mathbf{r}''} - 4q^4 \left[\frac{\mathbf{h}' + \mathbf{h}''}{\mathbf{r}' + \mathbf{r}''} \right]^2 \left[1 + \frac{i\mathbf{a}}{q^2} \frac{\mathbf{r}' + \mathbf{r}''}{\mathbf{h}' + \mathbf{h}''} \right]^{1/2} = 0. \quad (3)$$

A more detailed and rigorous consideration of the dynamics of surface waves on liquid surfaces or gas-liquid interfaces can be found in literature, see, e.g., Refs. 2, 4, 6, 7, and 21.

2.2 Scattering Geometry

Light interacting with a oscillating surface structure is scattered. The scattering geometry used in this work is shown in Fig. 1, where scattered light is observed in forward direction near refraction. This arrangement has been chosen due to signal and stability considerations [22] and differs from the more commonly employed scattering geometry, where the scattered light is observed close to the direction of the reflected beam. By the choice of the angle of incidence θ , resulting in a specific angle α of the refracted light, and the scattering angle θ_s , the scattering vector $\vec{q} = \vec{k}'_1 - \vec{k}'_s$ is determined and by this the wave vector and frequency of the observed surface vibration mode. Here, \vec{k}'_1 and \vec{k}'_s denote the projections of the wave vectors of the refracted (\vec{k}_1) and scattered light (\vec{k}_s) in the surface plane, respectively. For the observation of scattered light within the irradiation plane and assuming elastic scattering (i.e. $k_1 \cong k_s$), the modulus of the scattering vector is obtained as

$$q = |\vec{k}'_1 - \vec{k}'_s| \cong 2k_1 \sin(\theta_s / 2) \cos(\alpha - \theta_s / 2) = \frac{4pn}{\lambda_0} \sin(\theta_s / 2) \cos(\alpha - \theta_s / 2), \quad (4)$$

where n is the fluid refractive index and λ_0 the laser wavelength in vacuo.

2.3 Correlation Technique

In light scattering experiments the surface oscillations described result in a temporal modulation of the scattered light intensity, which contains information on the dynamics of the surface. Information about this processes can be derived by a temporal analysis of the scattered light using photon correlation spectroscopy (PCS). For heterodyne conditions,

where the scattered light is superimposed with coherent reference light, e.g., with stray light from the windows of the measuring cell, the time correlation function for the analysis of surface fluctuations is described by [21]

$$G^{(2)}(\mathbf{t}) = A + B \cos(\mathbf{w}\mathbf{t}) \exp(-\mathbf{t}/\mathbf{t}_c), \quad (5)$$

where the correlation time \mathbf{t}_c and the frequency \mathbf{w} are identical with the time decay behavior ($G = 1/\mathbf{t}_c$) and the frequency of the surface oscillations. A and B are experimental constants.

The correlation function can thus be used for the evaluation of the desired properties surface tension and viscosity, see Eq. (2) for a first order approximation.

3. EXPERIMENTAL

The experimental set-up used is shown in Fig. 2. A frequency-doubled continuous-wave Nd:YVO₄-laser operated in a single mode with a wavelength of $\lambda_0 = 532$ nm serves as a light source. The laser power was about 250 mW when working at temperatures $T < 300$ K and somewhat lower for temperatures above. For the observation of light scattered by surface waves the optical path has to be aligned in a way that the laser beam and the direction of detection intersect on the liquid-vapor interface in the measurement cell. For large scattering intensities from the vapor-liquid interface scattered reference light from the cell windows is not sufficient to realize heterodyne conditions. Here, an additional reference beam is added. To this end, part of the incident laser light is splitted by a glass plate and superimposed to the scattered light behind the sample cell. The time-dependent intensity of the scattered light is

detected by two photomultiplier tubes (PMTs) operated in cross-correlation in order to suppress afterpulsing effects. The signals are amplified, discriminated, and fed to a digital correlator with 256 linearly spaced channels operated with a sample time down to 50 ns. Light scattered on the liquid-vapor interface is detected perpendicular to the surface plane, which means $\mathbf{Q}_S = \mathbf{d}$, see Fig. 1. For this arrangement, with the help of Snell's refraction law and simple trigonometric identities, the modulus of the scattering vector q can be deduced as a function of the easily accessible angle of incidence

$$q = \frac{2p}{I_0} \sin(\mathbf{Q}_E). \quad (6)$$

For the measurement of the angle of incidence \mathbf{Q}_E , the laser beam is first adjusted through the detection system consisting of two apertures (\varnothing 1 - 2 mm) at a distance of about 4 m. Then the laser beam is set to the desired angle. For the experiment the angle of incidence \mathbf{Q}_E was set between 3.0 ° and 4.5 ° and was measured with a high precision rotation table. The error in the angle measurement has been determined to be approximately ± 0.005 °, which results in a maximum uncertainty of less than 1% for the desired thermophysical properties.

According to the specification of the manufacturer (Merck GmbH, Darmstadt) the toluene sample was of spectroscopic grade (Uvasol®) with a minimum purity of 99.9% and was used without further purification. For the present measurements, the sample was filled from the liquid phase into an evacuated cylindrical pressure vessel (diameter 70 mm; volume 150 cm³) equipped with two quartz windows (Herasil I, diameter 30 mm x 30 mm). The temperature regulation of the cell surrounded by an insulating housing was realized with

electrical heating. For temperatures below room temperature the insulating housing was cooled down to about 10 K below the desired temperature in the sample cell by a lab-thermostat. The temperature of the cell was measured with two calibrated Pt-100 Ω resistance probes, integrated into the main body of the vessel, with a resolution of 0.25 mK using an AC bridge (Paar, MKT 100). The accuracy of the absolute temperature measurement was better than ± 0.015 K. The temperature stability during an experimental run was better than ± 0.001 K. For each temperature, at least six measurements at different angles of incidence were performed, where the laser was irradiated from either side with respect to the axis of observation in order to check for a possible misalignment. The measurement times for a single run were typically of the order of ten minutes down to few seconds for the highest temperatures in this study.

4. MEASUREMENT EXAMPLE AND DATA EVALUATION

Fig. 3 shows an example of a correlation function as it was obtained by scattering on a liquid-vapor interface of toluene under saturation conditions at a temperature of 303.15 K. The experimental correlation function, Eq. (5), has to be evaluated for the central quantities ω and t_c , which may be done directly by a standard nonlinear fit in which the squared sum of residuals is to be minimized. Within the interesting fit range no systematic deviations can be observed. This is illustrated in the example of the residual plot in Fig. 3 and was confirmed for all measurements. The fit to the experimental correlation function results in a frequency of $\omega = 2.6035 \times 10^6 \text{ rad} \cdot \text{s}^{-1} \pm 0.03\%$ and a decay time of $t_c = 2.701 \times 10^{-6} \text{ s} \pm 0.2\%$. The standard errors obtained from the fit may be compared with the deviations obtained from fits

to various fit intervals, varying the first channel included in the fit in a range up to $0.5t_c$ and the last channel in a range starting at $2t_c$. With this procedure, the standard deviations of these individual fits are 0.04% for ω and 0.3% for t_c . It should be noted here that either value is only indicative for the order of magnitude of the uncertainty that is related with the determination of the frequency and decay time of the measured correlation function. It is obvious that the error in the determination of the frequency is one order of magnitude smaller than that of the decay time.

The evaluation of the experimental data for the desired quantities viscosity and surface tension has always performed on basis of a full solution of the dispersion relation, Eq. (3). The necessity for this approach is illustrated in Fig. 4, where experimental values for the damping G ($=1/t_c$) and frequency ω for a vapor-liquid interface of toluene at temperatures of 303.15 K and 373.15 K are shown over a wide range of wave numbers. The drawn and dashed lines indicate the theoretical variations obtained by an exact numerical solution of the dispersion equation Eq. (3) and by a derivation according to the first order approximation Eq. (2), respectively. For both calculations data for the density of the liquid and vapor phase have been adopted from the equation of state (EOS) of Goodwin [23]. Furthermore, the dynamic viscosity of the liquid phase was calculated from a correlation as given by Nieto de Castro and Santos [24], which is capable to describe the most experimental data sets for toluene within its own stated accuracy, while data for the dynamic viscosity of the vapor phase are calculated theoretically according to a method given in Refs. 25 and 26. Finally, an estimation method for nonpolar liquids as described in Ref. 26 was used to compute the surface tension of toluene. As it can be seen from Fig. 4, with the exception of the highest wave numbers investigated in

this work at a temperature of 303.15 K, where the scattered signal was weak and only a poor accuracy could be achieved, excellent agreement can be found for the measured values of ω and G with the theoretical calculation based on an exact solution of the dispersion equation. In contrast to this, an increasing difference with increasing wave numbers is observable in respect of the first order approximation. This behavior makes clear, especially for the relatively high wave numbers studied in this work, that a reliable determination of surface tension and viscosity is only possible by an exact numerical solution of the dispersion equation Eq. (3), where the frequency ω , damping G , and the modulus of the scattering vector q are used as input values. It should be emphasized that for the determination of surface tension and viscosity we always used wave numbers in range of from about 0.6 to $1.1 \times 10^6 \text{ m}^{-1}$. The lower limit was chosen so that instrumental broadening effects are negligible, while the limitation to q -values smaller than $1.1 \times 10^6 \text{ m}^{-1}$ is due to a weak scattering signal, as already mentioned above.

5. EXPERIMENTAL RESULTS AND DATA CORRELATION

The quantity directly accessible in surface light scattering experiments is the ratio $\tilde{S} = S / (r' + r'')$ of the surface tension S to the sum of the densities of the liquid and vapor phase. Similarly, also the direct quantity \tilde{n} obtained for the viscosity is determined by both vapor and liquid properties, i.e. $\tilde{n} = (h' + h'') / (r' + r'')$, where h' and h'' are the dynamic viscosities of the liquid and vapor phase, respectively. If appropriate reference data for the quantities of the vapor phase are not available the approximation $\tilde{n} \approx n'$ can be used, which relies on the neglect of vapor properties as compared with the respective liquid quantities, and

thus yields an approximate kinematic liquid viscosity. An estimation based on our experimental values for the quantity \tilde{n} , data for the liquid and vapor density from the work of Goodwin [23], and on theoretically calculated values for the dynamic viscosity of the vapor phase according to the method given in Refs. 25 and 26 indicates that for toluene the approximation would result in a systematic deviation from the exact kinematic viscosity value of about + 1% for the lowest temperatures studied in this work, see Fig. 5. With increasing temperature the systematic error caused by neglecting the influence of the vapor phase would increase up to + 3%. In the present work, however, data obtained for \tilde{n} and \tilde{S} by an exact solution of the equation of dispersion for surface waves, see Eq. (3), have been combined with theoretically calculated data for the dynamic viscosity of the vapor phase, see Refs. 25 and 26, and with density data for both phases from the EOS of Goodwin [23], to get the information about the surface tension S and liquid kinematic viscosity n' .

The results for the desired quantities from surface light scattering are summarized in Table I. The listed data are average values of at least six independent measurements with different angles of incidence Q_E . Also listed in Table I are the quantity \tilde{n} obtained for the viscosity directly from the experiment, and the values from literature used for data evaluation as described above. With the approach given in Refs. 25 and 26 the vapor viscosity data can normally be predicted within $\pm 10\%$ for the temperature range studied in this work which does not have any appreciable influence on the total accuracy of better than 1% for the liquid kinematic viscosity. This estimate for the uncertainty of our viscosity values is obtained from

$$\Delta \mathbf{n}' \approx \left[\left[\frac{\mathbf{r}' + \mathbf{r}''}{\mathbf{r}'} S_{\tilde{n}} \right]^2 + \left[\frac{1}{\mathbf{r}'} \Delta \mathbf{h}'' \right]^2 + \left[\frac{\mathbf{h}'' - \tilde{n} \mathbf{r}''}{\mathbf{r}'^2} \Delta \mathbf{r}' \right]^2 + \left[\frac{\tilde{n}}{\mathbf{r}'} \Delta \mathbf{r}'' \right]^2 \right]^{1/2}, \quad (7)$$

based both on the standard deviation $S_{\tilde{n}}$ of the measurement values and on the uncertainty of the reference data needed for the determination of true liquid kinematic viscosity from the direct observable \tilde{n} . In detail, for the relative uncertainty of the vapor viscosity $\Delta \mathbf{h}''/\mathbf{h}''$, liquid density $\Delta \mathbf{r}'/\mathbf{r}'$, and vapor density $\Delta \mathbf{r}''/\mathbf{r}''$ values of 10%, 1%, and 1%, respectively, have been assumed. As it is true for many DLS applications [27], the standard deviation of individual measurements may be considered as a reasonable measure for the experimental uncertainty. In all instances, the value for $S_{\tilde{n}}$ was below 1%, which is mainly determined by the uncertainty of the angle measurement. The relative overall maximum uncertainty $\Delta \mathbf{n}'/\mathbf{n}'$ of our values for the liquid kinematic viscosity as estimated by Eq. (7) is displayed in Fig. 6. Here, the individual contributions of Eq. (7) related to $S_{\tilde{n}}$, $\Delta \mathbf{h}''$, $\Delta \mathbf{r}'$, and $\Delta \mathbf{r}''$ are shown relative to the values of \mathbf{n}' . As it can be seen from Fig. 6, for the whole temperature range studied in this work the uncertainties in the used reference data have comparatively small influence on the final results for liquid viscosities, so that for this quantity an overall maximum uncertainty of better than 1% could be established. In a similar way the uncertainty for the surface tension may be estimated. For the whole temperature range studied the standard deviation $S_{\tilde{s}}$ of individual measurements was in most cases below $\pm 0.5\%$, and although the accuracy of density data is of course far better than those of vapor viscosity data, some uncertainty is also introduced through the limited accuracy of the available density data.

Yet in combination, a value of better than 1% may be regarded as a fair estimate for the total uncertainty of the surface tension.

For the whole temperature range studied in the present investigation, a modified Andrade-type equation, which in its simple form is commonly chosen to represent the dynamic viscosity at least over a limited temperature range, was used in the form

$$\boldsymbol{n}' = \boldsymbol{n}'_0 \exp[\boldsymbol{n}'_1 T^{-1} + \boldsymbol{n}'_2 T] \quad (8)$$

in order to represent our experimental kinematic viscosity data for toluene, where T is the temperature in Kelvin and the coefficients are given in Table II. Here, also the standard deviation of our data relative to those calculated by Eq. (8) is listed. It should be noted that the residuals of the experimental data from the fit are smaller than the standard deviation of the individual measurements. The experimental data for the surface tension can be well represented by a linear equation of the form

$$\boldsymbol{s} = \boldsymbol{s}_0 + \boldsymbol{s}_1 T, \quad (9)$$

where the fit parameters \boldsymbol{s}_0 and \boldsymbol{s}_1 are given in Table III. The present correlation represents the experimental values of the surface tension with a root mean square deviation of about 0.2%.

6. COMPARISON WITH LITERATURE DATA

In Fig. 7 our values for the kinematic viscosity of toluene under saturation are shown in comparison to available literature data from the last twenty years. Deviations between our results from surface light scattering and the reference values are plotted using our correlation Eq. (8) as a basis. Data for the viscosity included in Fig. 7 comprise measurements of Medani and Hasan [28] performed by a rolling ball viscometer, a method where it is questionable if low uncertainties can be achieved, and data by Dymond and Robertson [29] obtained with a capillary viscometer with a stated uncertainty of $\pm 0.5\%$. The measurements by Byers and Williams [30], Gonçalves et al. [31], and Kaiser et al. [32] were performed by Ubbelohde capillary viscometers with claimed uncertainties of 0.3%, 0.3%, and around 1%, respectively. These data sets and the compilation by Vargaftik [33] refer to atmospheric pressure, the deviation from saturation values is negligible for the whole temperature range in this study, where the maximum saturation pressure is 0.1 MPa. Finally, beside the already mentioned correlation by Nieto de Castro and Santos [24], which describes the most recently reported experimental data sets within its own stated accuracy, a correlation by Krall et al. [34] has been included, which is based on experimental values from an oscillating-disc viscometer with a stated experimental accuracy of $\pm 0.5\%$. For the computation of these correlations and the conversion of the data in Refs. 28 and 33 from dynamic to kinematic viscosity density data from the equation of state by Goodwin [23] have been employed. Figure 7 shows an excellent agreement between our data from surface light scattering and those given by Gonçalves et al. [31] and by Dymond and Robertson [29]. Within the combined uncertainties this statement also holds for the data given by Dymond and Robertson [29] and for the correlation by Krall

et al. [34]. Furthermore, particularly good agreement with an average deviation of 0.34% can be found between the fit of our data and the compilation by Vargaftik [33]. For the viscosity correlation given by Nieto de Castro and Santos [24] and the experimental data by Kaiser et al. [32] at low temperatures ($T < 275$ K) a positive deviation from our values can be recognized, which slightly exceeds the combined uncertainties. The fundamental similar behavior of both data sets at low temperatures may reflect that the correlation of Nieto de Castro and Santos [24] is only based on the experimental data of Kaiser et al. [32]. While for temperatures above 275 K good agreement can be found between our values and the correlation given by Nieto de Castro and Santos [24], for the data given by Kaiser et al. [32] there a deviation from our data can be recognized which exceeds the combined uncertainties. It should be noted, as our experimental values were limited to a maximum temperature of 383.15 K, the regression Eq. (8) takes the character of an extrapolation at higher temperatures.

Values for the surface tension of toluene from surface light scattering are plotted in Fig. 8 together with available reference data extending over the past century. For data comparison only references are taken into account which include at least three surface tension values at different temperatures. All experimental data displayed in Fig. 8 by symbols are based on the capillary rise method [35-41], with the exception of the data sets by Donaldson and Quayle [42] and Buehler et al. [43], which are determined by the maximum bubble pressure method. Furthermore, all experimental data refer to atmospheric pressure, with the exception of the data by Morino [36], which were obtained at saturation conditions. While in Fig. 8 the depicted correlation of Körösi and Kováts [44] is based on their own experimental values

from the capillary rise method, the correlation given by Jasper [45] is based on the work of Donaldson and Quayle [42]. The surface tension correlation given by Bonnet and Pike [46] is based on 58 experimental data points collected from literature. Finally, values from the data collection of Vargaftik [33] and an estimation by Reid et al. [26] as already mentioned above, are included in Fig. 8. Here, for the few data sets which explicitly give a statement for the uncertainty a value between about 0.5% and 1% can be found. As it can be seen from the deviation plot of Fig. 8, where the deviations between our results and the reference values are plotted using our correlation Eq. (9) as a basis, the maximal differences between the different data sets are partly larger than 8%. The experimental data sets seem to form two bands, one clearly above and one clearly below our values from surface light scattering. In contrast to this behavior, for the whole temperature range investigated in the present study good agreement between our values and the prediction of Reid et al. [26] can be found. Comparing our data with the recommended values of Jasper [45] a decreasing deviation can be observed with increasing temperature. For these values the differences at low temperatures are outside the combined uncertainty, while good agreement can be found for temperatures $T > 330$ K. The same statement also holds for the work of Körösi and Kováts [44]. Summarizing it seems that the surface tension of toluene is not known more accurately than $\pm 2\%$. It is not surprising that there are large discrepancies in the given values for surface tension as the determination of this property may be affected by two factors which may not be easily controlled experimentally. First, values for surface tension are extremely susceptible to contamination. Second, if surface tension is measured for liquid-air systems as in most cases cited above the surface temperature may be somewhat below the temperature in the bulk of the fluid. An influence of this error,

however, can be excluded for the present investigation, which has been carried out under saturation conditions in thermodynamic equilibrium.

7. CONCLUSIONS

Our investigations on a horizontal liquid-vapor interface of toluene under saturation conditions have shown that the surface light scattering technique can be utilized for an efficient and reliable determination of liquid kinematic viscosity and surface tension of transparent fluids. With the help of reference data for the liquid and vapor density and theoretically calculated data for the dynamic viscosity of the vapor phase - yet without any calibration procedure - an overall uncertainty of $\pm 1\%$ could be achieved for both properties of interest. Measurements have been performed over a temperature range from 263 K to 383 K. For the kinematic viscosity the agreement with recently literature data can be regarded as fully satisfactory. For the surface tension of toluene a comparison of reference data covering the past century points to large differences of up to 8%, where our data from surface light scattering seems to form the center. With respect to recommended values for the surface tension of toluene a maximum deviation of about - 2.5% can be found for the lowest temperatures studied in this work. An improvement of the situation for the surface tension of toluene requires more accurate measurements under well defined conditions.

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Table I: Viscosity and surface tension of toluene under saturation conditions.

T , K	$\tilde{\eta}$, $\text{mm}^2\cdot\text{s}^{-1}$	h'' , $\mu\text{Pa}\cdot\text{s}$	r' , $\text{kg}\cdot\text{m}^{-3}$	r'' , $\text{kg}\cdot\text{m}^{-3}$	n' , $\text{mm}^2\cdot\text{s}^{-1}$	s , $\text{mN}\cdot\text{m}^{-1}$
263.15	0.9965	6.06	892.3	0.02	0.9897	31.02
273.15	0.8710	6.29	883.7	0.04	0.8639	30.06
283.15	0.7707	6.52	874.9	0.07	0.7633	28.92
293.15	0.6870	6.76	866.1	0.11	0.6793	27.94
303.15	0.6180	6.99	857.1	0.18	0.6100	26.79
313.15	0.5606	7.23	848.0	0.28	0.5523	25.61
323.15	0.5104	7.46	838.8	0.42	0.5018	24.50
333.15	0.4710	7.71	829.5	0.62	0.4621	23.42
343.15	0.4343	7.96	820.0	0.89	0.4251	22.31
353.15	0.4052	8.21	810.3	1.24	0.3957	21.18
363.15	0.3766	8.47	800.5	1.70	0.3668	20.19
373.15	0.3543	8.74	790.4	2.27	0.3443	19.10
383.15	0.3332	9.01	780.2	2.99	0.3229	17.99

Table II: Coefficients of Eq. (8).

$n'_0, \text{mm}^2\cdot\text{s}^{-1}$	0.013115
n'_1, K	1057.46
n'_2, K^{-1}	0.0011597
rms, %	0.14
T -range, K	263-383

Table III: Coefficients of Eq. (9).

$s_0, \text{mN}\cdot\text{m}^{-1}$	59.933
$s_1, \text{mN}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$	-0.10952
rms, %	0.21
T -range, K	263-383

FIGURE CAPTIONS

Fig. 1 Scattering geometry.

Fig. 2 Experimental setup: optical and electronic arrangement.

Fig. 3 Fit to a normalized experimental correlation function and residuals.

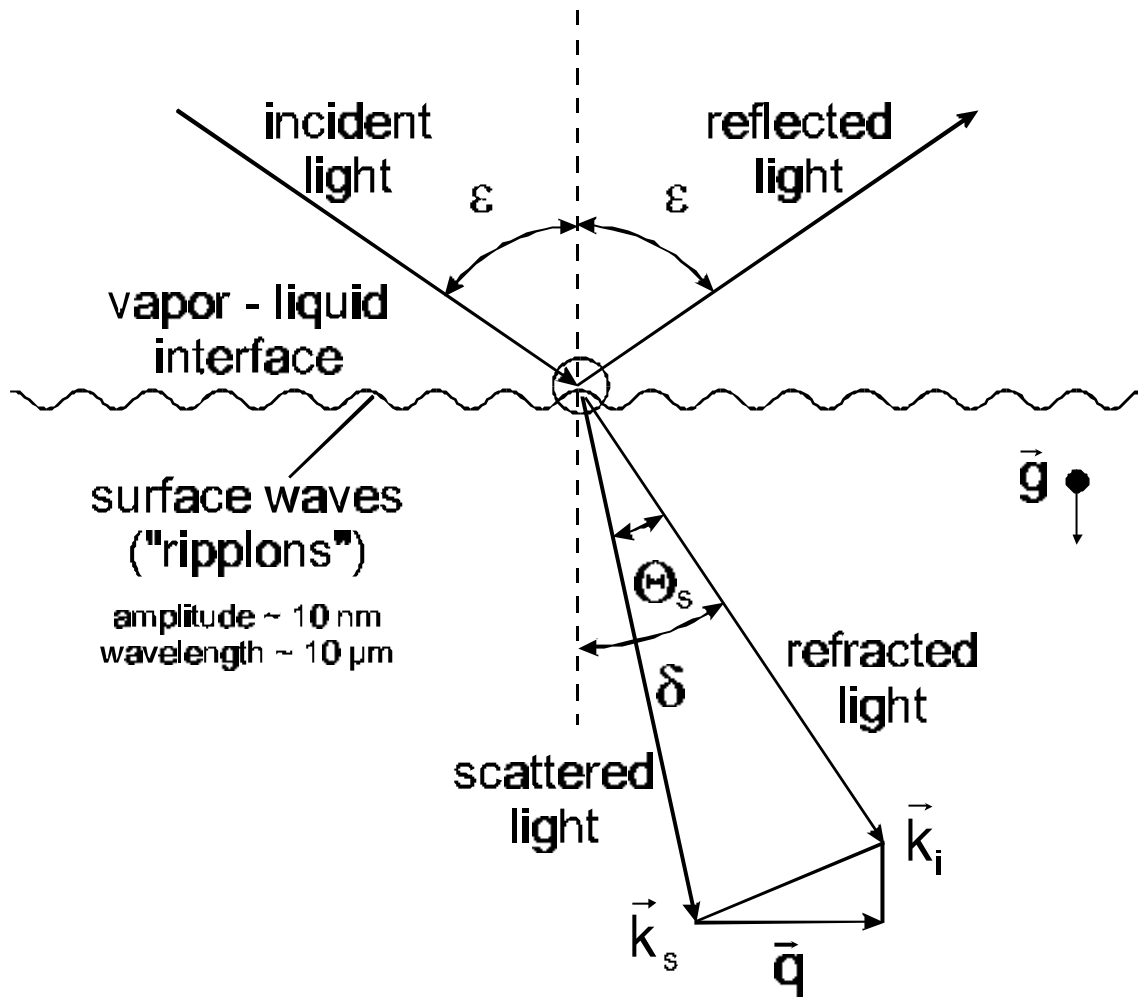
Fig. 4 Frequency ω and damping G of surface waves on a horizontal gas-liquid interface of toluene under saturation conditions in depending on the wave vector q at a temperature of 303.15 K and 373.15 K: (■) experimental values from surface light scattering; (—) theoretically calculated values by a numerical solution of the dispersion equation Eq. (3); (— —) theoretically calculated values by a first order approximation Eq. (2).

Fig. 5 Deviation between the direct accessible quantity $\tilde{\eta}$ for the viscosity from surface light scattering and the kinematic viscosity ν' of liquid toluene under saturation conditions.

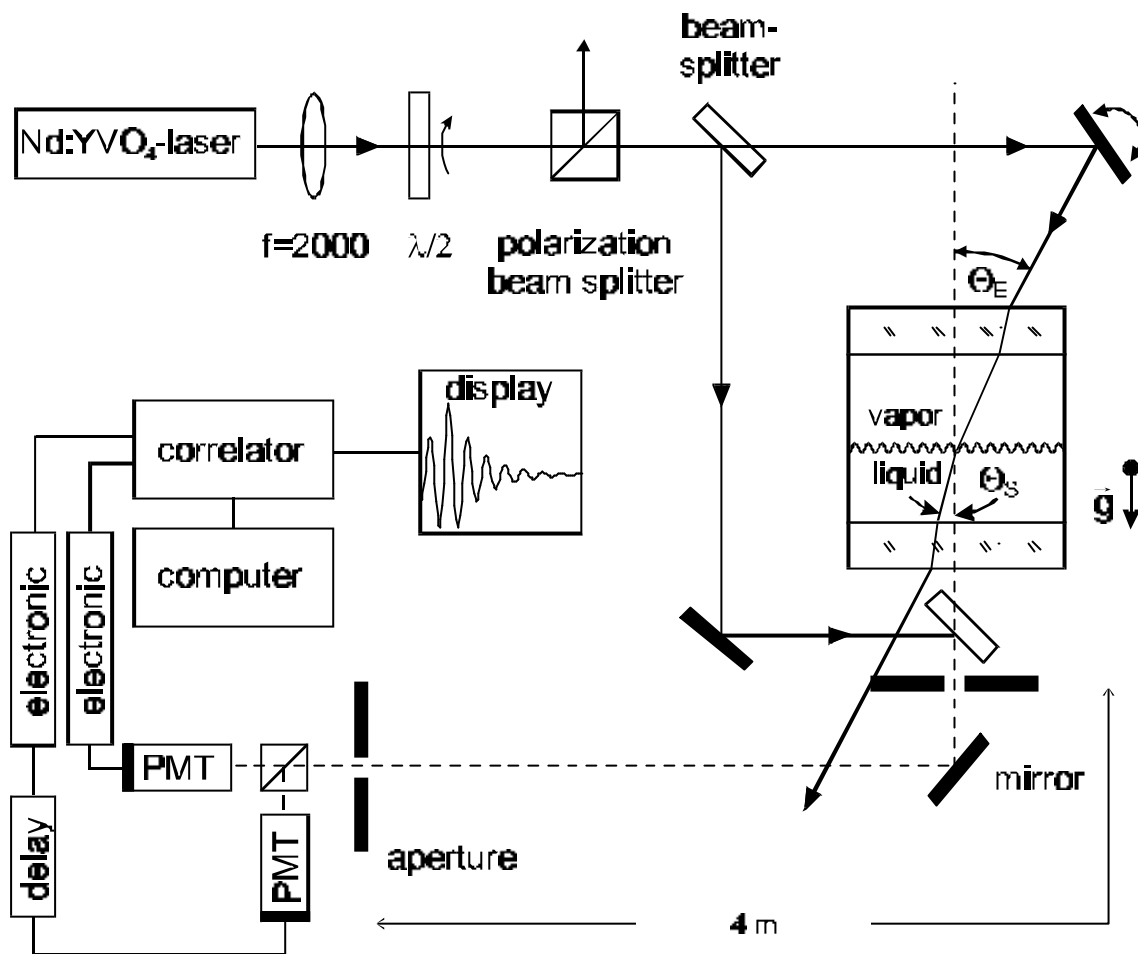
Fig. 6 Estimated overall maximum uncertainty for the liquid kinematic viscosity and individual contributions to that value.

Fig. 7 Kinematic viscosity of liquid toluene under saturation conditions from surface light scattering in comparison to literature.

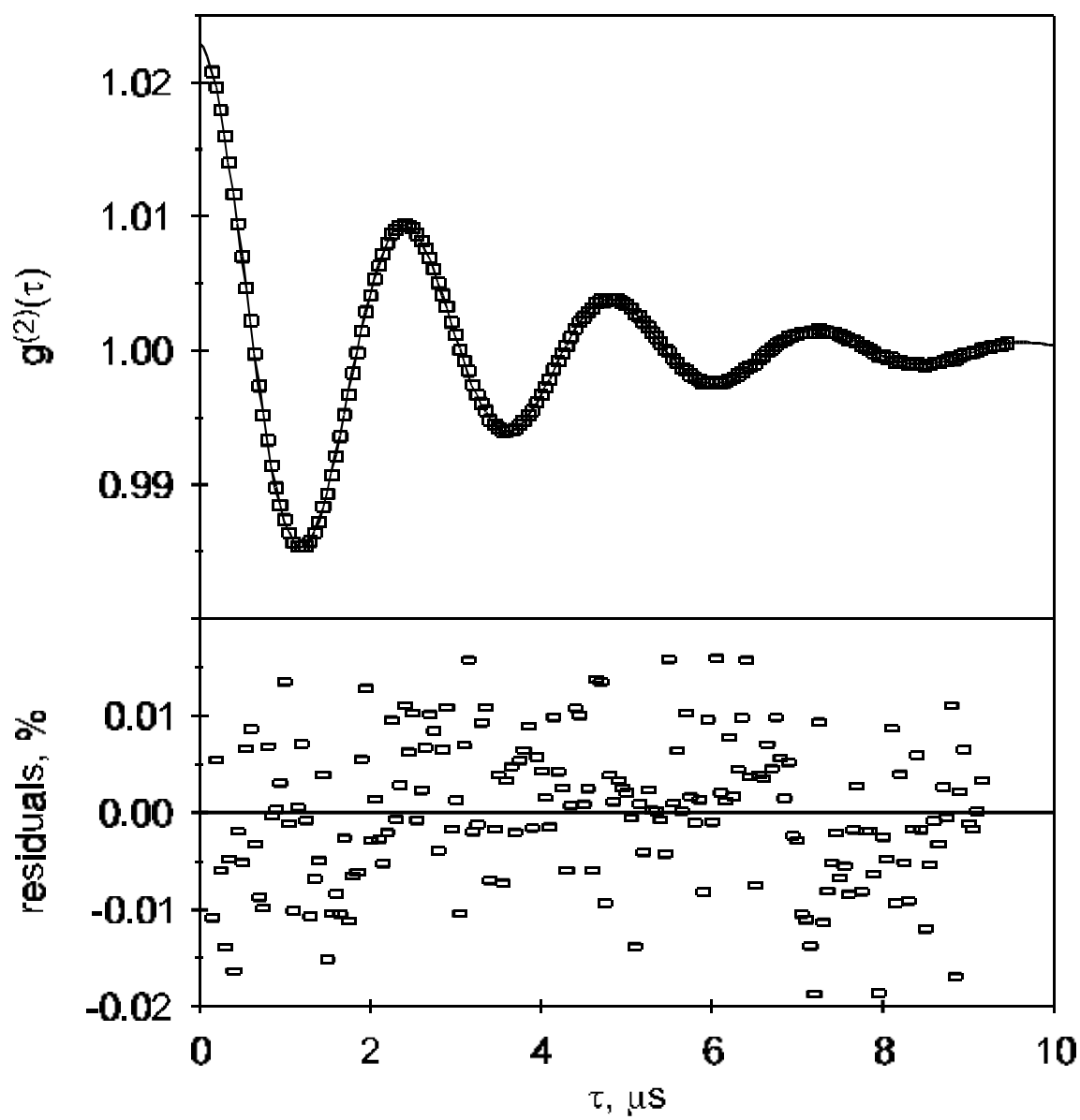
Fig. 8 Surface tension of toluene under saturation conditions from surface light scattering in comparison to literature.



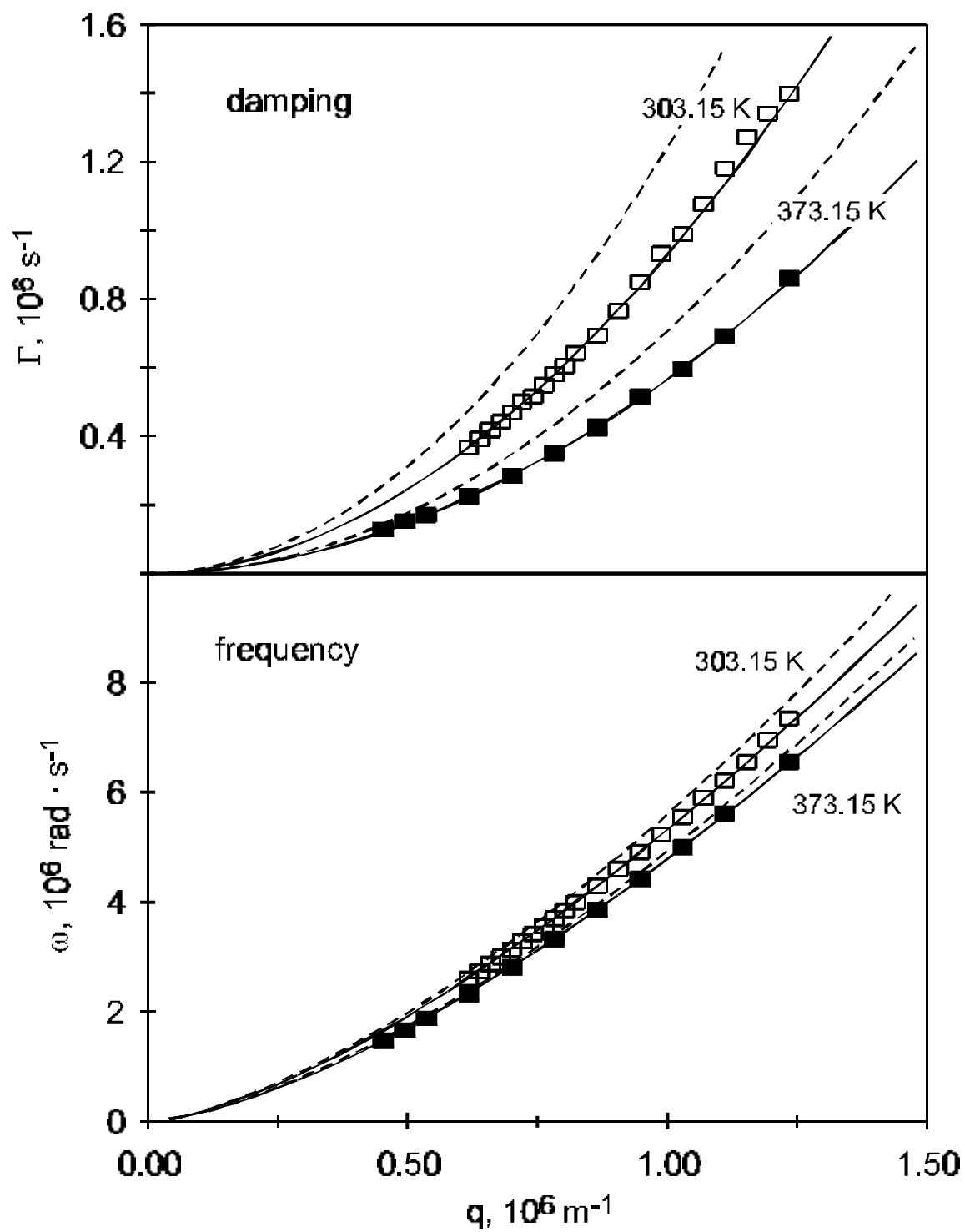
Fröba and Leipertz, Fig. 1



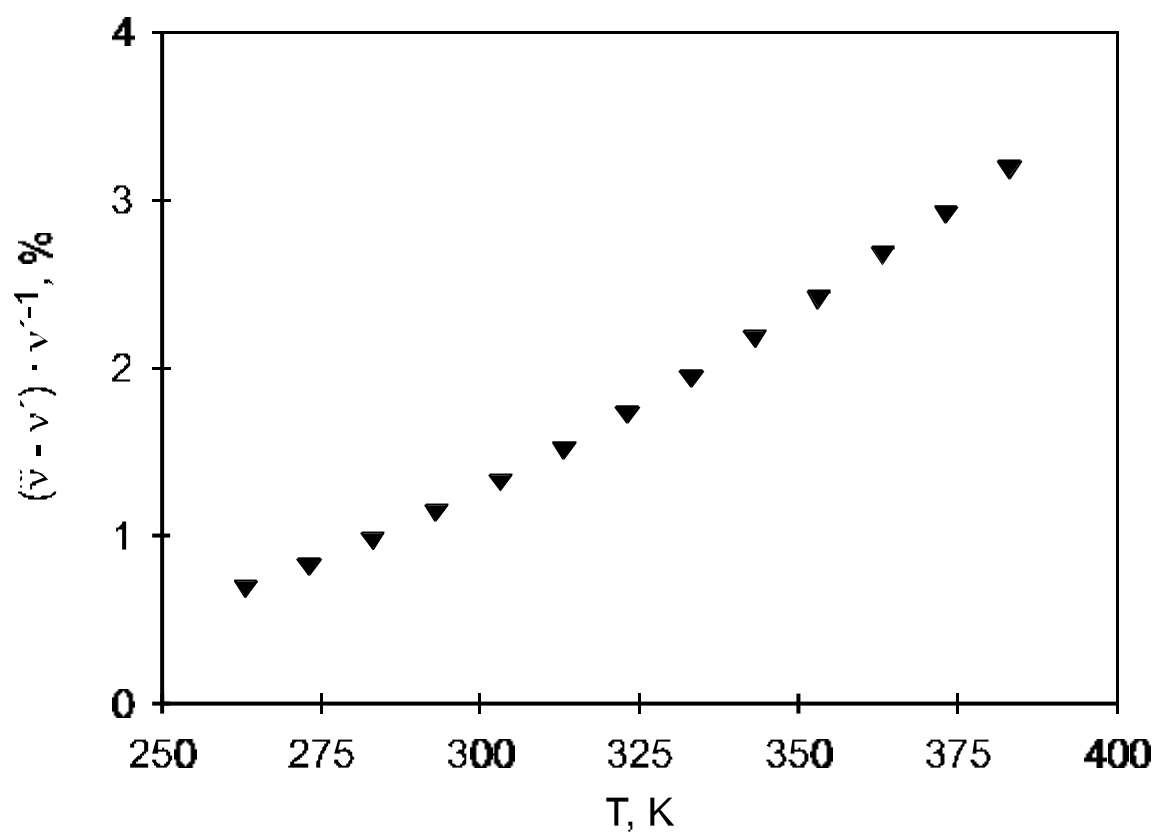
Fröba and Leipertz, Fig. 2



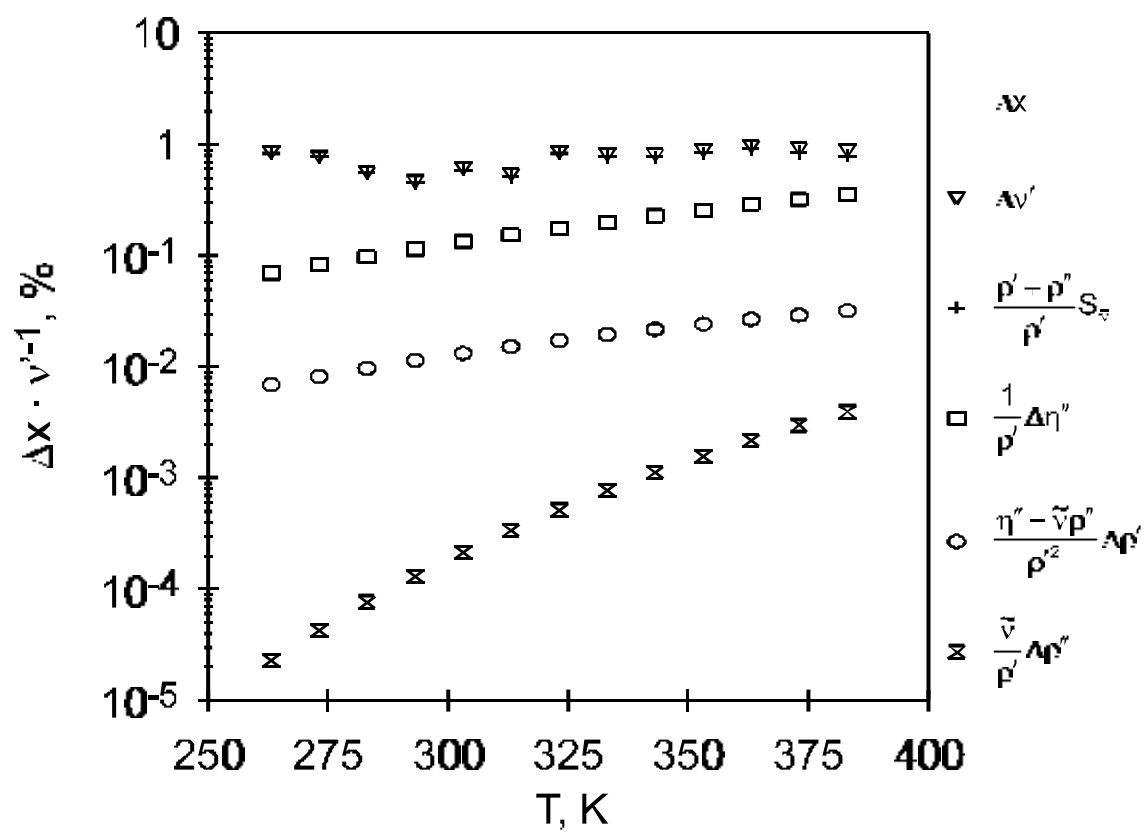
Fröba and Leipertz, Fig. 3



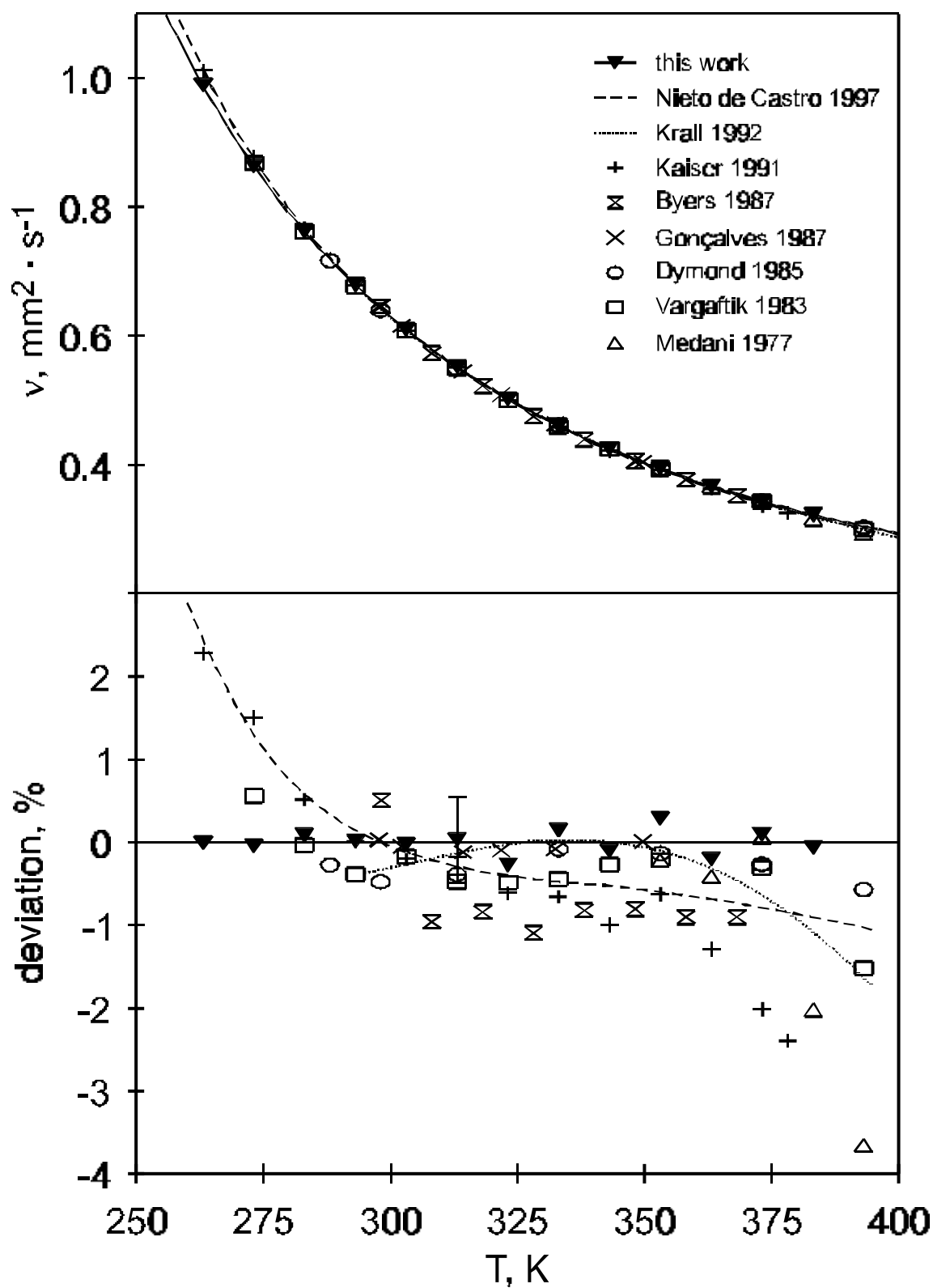
Fröba and Leipertz, Fig. 4



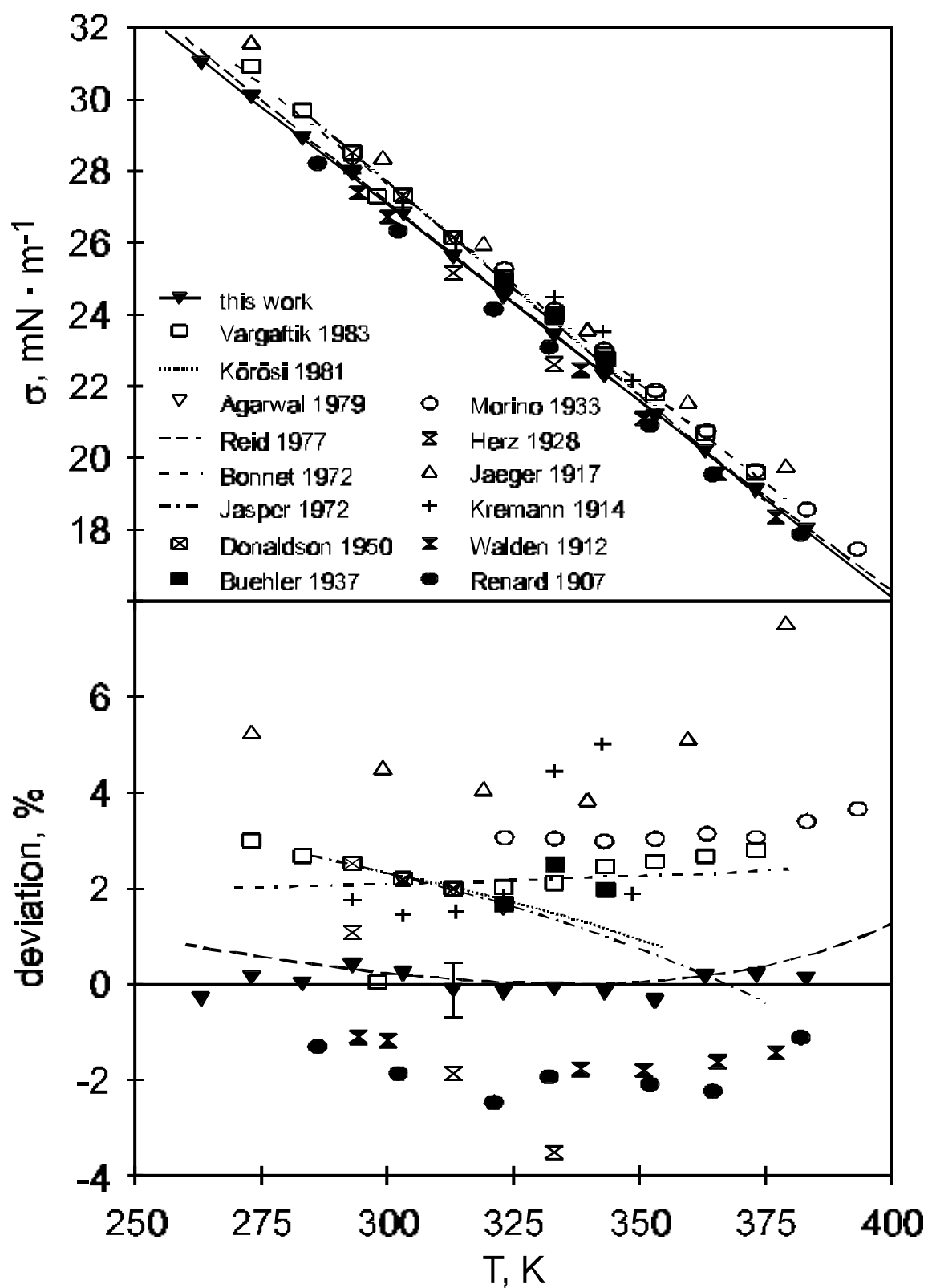
Fröba and Leipertz, Fig. 5



Fröba and Leipertz, Fig. 6



Fröba and Leipertz, Fig. 7



Fröba and Leipertz, Fig. 8